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SOOT FORMATION IN PURELY-CURVED PREMIXED FLAMES AND LAMINAR FLAME SPEEDS OF SOOT-FORMING FLAMES

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Task Objectives

This is a collaborative research project of Professors C.-J. Sung (Case Western Reserve University) and H. Wang (University of Delaware). The research aims to gain fundamental understanding of the structure and dynamics of sooting premixed flames in simple, well-defined, one-dimensional, burner-supported, cylindrical and spherical flow fields. The proposed program consists of the following four major components: (1) experimental determination of the fundamental sooting limits in the absence of conductive heat loss to the burner; (2) experimental determination of the fundamental laminar flame speed of soot-forming flames; (3) experimental mapping of sooting structure; and (4) detailed modeling of sooting flame propagation and soot formation. The work in components 1 and 2 demonstrate a novel experimental approach under microgravity and generate fundamental flame and soot data that are suitable for rigorous comparison with detailed numerical simulation. This experimental approach eliminates the ambiguities associated with the boundary conditions in previous soot laminar premixed flame experiments and simulation. Combined with components 3 and 4 of the proposed work, we develop a soot research platform which is unique and complementary to the existing soot research tools and facilities.

INTRODUCTION

The research addressed here is a collaborative project between University of Delaware and Case Western Reserve University. There are two basic and related scientific objectives. First, we wish to demonstrate the suitability of spherical/cylindrical, laminar, premixed flames in the fundamental study of the chemical and physical processes of soot formation. Our reasoning is that the flame standoff distance in spherical/cylindrical flames under microgravity can be substantially larger than that in a flat burner-stabilized flame. Therefore the spherical/cylindrical flame is expected to give better spatial resolution to probe the soot inception and growth chemistry than flat flames. Second, we wish to examine the feasibility of determining the laminar flame speed of soot forming flames. Our basic assumption is that under the "adiabatic" condition (in the absence of conductive heat loss), the amount and dynamics of soot formed in the flame is unique for a given fuel/air mixture. The laminar flame speed can be rigorously defined as long as the radiative heat loss can be determined. This laminar flame speed characterizes the flame soot formation and dynamics in addition to the heat release rate.

* Moved to the University of Southern California prior to the completion of the project.

The research involves two integral parts: experiments of spherical and cylindrical sooting flames in microgravity (CWRU), and the computational counterpart (UD) that aims to simulate sooting laminar flames, and the sooting limits of near adiabatic flames. The computational work is described in this report, followed by a summary of the accomplishments achieved to date. Details of the microgravity experiments will be discussed in a separate, final report prepared by the co-PI, Professor C.-J. Sung of CWRU. Here only a brief discussion of these experiments will be given.

MICROGRAVITY EXPERIMENT

This work focuses on the study of sooting flames using spherical and cylindrical flame configurations in microgravity. These two configurations are preferable to the popular twin counterflow flame method because they prevent the sooting non-uniformity as found in the counterflow geometry. The use of a microgravity spherical flame also allows the simple determination of the unstretched burning velocity by varying the mass flow rate of the reactants and measuring the resulting flame radius. Since the flame location must be accurately known, a simple and compact optical technique first was developed and tested at CWRU. The technique developed was shown to be capable of accurate burning velocity measurements for sooting flames. This work has been reported in publications [1-5].

THE ROLE OF IONS IN SOOT MASS GROWTH

In sooting flames, a large fraction of the soot particles may be charged due to thermal ionization. The density of charged particles is expected to increase exponentially with an increase in flame temperature and particle size. Due to charge interactions, the coagulation of charged-neutral particles can be notably enhanced. Furthermore, the presence of charge on the particles may also affect the surface reaction rate. Although the role of ions in soot formation has been extensively discussed, the influence of charged particles in soot mass growth has not been quantitatively examined by considering the detailed particle size distribution function. We carried out a detailed numerical simulation study with the goal to assessing the role of charged particles in soot mass growth. The results show that at 2000 K, as many as 50% of the particles are charged. A rigorous consideration of coagulation enhancement due to charge interactions gives, however, little difference in the predicted soot volume fraction. This result allowed us to conclude that as far as the simulation of soot formation is concerned, the omission of thermal ionization does not lead to significant differences in the predicted growth of soot particles. Thermal ionization would play a role in the growth of soot particles if the surface reactions between charged particles and gaseous molecules were enhanced. Nevertheless, the uncertainty associated with the omission of thermal ionization is significantly smaller than the uncertainties in the kinetics of particle inception and surface growth in most laboratory flames. This work was reported in publication [6].

SIMULATION OF DETAILED SOOT PARTICLE SIZE DISTRIBUTION FUNCTION

In a recent studies [7, 8] detailed soot particle size distribution functions (PSDFs) were measured using a nano scanning mobility particle sizer. It was found that the PSDFs could be bimodal, depending on the strength of particle nucleation in the post flame. The characteristic bimodal PSDF shape is expected to be pertinent to flames with maximum flame temperature $< \sim 1700$ K. The

bimodality poses a challenge for accurate prediction of the intensity of soot radiation, which is greatly influenced by soot surface area and thus the shape of the PSDF. Moreover, the existence of the bimodal distribution provides a stringent validation test for the soot model, since the bimodal behavior affects the rate of soot mass growth and thus the soot yield. For this reason, we carried out a detailed numerical simulation using a recently developed stochastic method. The results show that the model predicts well the shapes of the measured PSDFs, although the particle size was somewhat under-predicted. The work was reported in publication [9].

THE ROLE OF FERROCENE IN SOOT FORMATION

It is known that while ferrocene suppresses soot formation in diffusion flames, it promotes soot formation in premixed flames. The consensus appears to be that in premixed flames iron or iron oxide would nucleate early into particles, which provide a substrate for carbon deposition. This phenomenon is viewed as soot nucleation induced by iron or iron oxide. In diffusion flames, however, the iron incorporation promotes the catalytic burnout of the carbon as soot transverse through the oxidizing region of the flame. In this study, we examined the critical sooting limits of premixed ethylene flames with and without ferrocene doping and found that the critical C/O ratios follows a characteristic, inverted bell shape dependence. The doped flames have smaller critical C/O ratio, indicating that the soot inception is indeed promoted by ferrocene. Computationally, the possible influence of the cyclopentadienyl resulting from ferrocene decomposition is examined. The computational results reproduced well the inverted bell shape dependence. The concentration of cyclopentadienyl radicals due to ferrocene pyrolysis was found to be too small to influence the PAH concentration and thus the homogeneous nucleation of soot. This result further supports the notion that, in doped flames soot nucleation is induced by iron or iron oxide particles. This work was reported in publication [10].

ACCOMPLISHMENTS

The research resulted in a novel approach to measure the laminar burning velocity of sooting flames, and a much improved understanding of soot nucleation and mass growth in hydrocarbon flames.

RESEARCH SIGNIFICANCE

The improved understanding for the sooting processes in hydrocarbon flames provides sound scientific basis for rational design of many combustors, including Diesel engines, gas-turbine and jet engines. The work is also of significance to developing rational strategies to reduced particulate air pollutants.

NEW TECHNOLOGIES DEVELOPED

None.

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